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WADC TECHNICAL REPORT 52-27

COATING OF STAINLESS- AND CARBON-STEEL FOIL

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DECEMBER 1951

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WRIGHT AIR DEVELOPMENT CENTER

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University of Illinois

December 1951

Power Plant Laboratory
Contract No. W33-038 ac-14520
E. O. No. R-506-67

Wright Air Development Center
Air Research and Development Command
United States Air Force
Wright-Patterson Air Force Base, Dayton, Ohio

McGregor & Werner, Inc., Wakefield, Mass.
Mar. 26, 1952 150

FOREWORD

This report, No. 59, was prepared by W. J. Plankenhorn and Dwight G. Bennett, at the University of Illinois in the Department of Ceramic Engineering under U. S. Air Force Contract No. W33-038 ac-14520. It covers the results of applying refractory ceramic coatings to .002 inch thick sheets of Inconel, Nichrome, 18-8 stainless-steel types 321 and 347 and carbon-steel foils. The technical phases of the contract are administered by the Power Plant Laboratory of the Wright Air Development Center, with Lt. Col. R. A. Jones acting as Project Engineer. It is identified as a project No. 1035 under E. O. No. R-506-67, **Ceramic Components for Aircraft Power Plants.**

ABSTRACT

Ceramic Coatings developed for application to alloy metals and other coatings developed for ingot iron and low carbon steel were successfully applied to specimens cut from .002 inch thick sheets of Inconel, Nichrome, 18-8 stainless-steel types 321 and 347 and to carbon steel, respectively. Scouring the metal surfaces with coating slip, followed by washing and drying, proved to be a satisfactory method for preparing the alloy metals for coating. A ten-minute anneal at 1800°F. improved the adherence of the coatings to the metal, particularly in the cases of the Inconel and Nichrome foils which had polished surfaces. Scouring and pickling were both found to be satisfactory methods for the preparation of the carbon steel foil. With extended heating at 1900°F. all uncoated alloy specimens showed some oxidation, as evidenced by a reduction in cross sectional area. Similar specimens, protected by refractory ceramic coatings, showed little or no reduction in metal thickness. Complete oxidation of the uncoated carbon steel foil occurred after 17 hours at a temperature of 1400°F. while coated specimens withstood a total of 64 hours at temperatures up to 1600°F.

PUBLICATION REVIEW

The publication of this report does not constitute approval by the Air Force of the findings or the conclusions contained therein. It is published primarily for the exchange and stimulation of ideas.

FOR THE COMMANDING GENERAL:

NORMAN C. APPOLD
Colonel, USAF
Chief, Power Plant Laboratory
Aeronautics Division

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I. INTRODUCTION

1. Statement of the Problem

Over a period of several years there has been an interest in ceramic coatings for application to metal foils. The earlier requests were for coatings which would provide radiation reflective surfaces to serve as an insulating barrier in an attempt to keep the heat away from the metal. More recently there has been an expressed desire to obtain coatings which would make it possible to use alloy metal foil at higher operating temperatures or to substitute less critical metals for those containing relatively large percentages of such alloying elements as are in short supply. Steps have accordingly been taken to adapt ceramic coatings developed at the University of Illinois to this particular application.

2. Historical

The sponsor, The Power Plant Laboratory, Wright Air Development Center, Wright-Patterson Air Force Base, Dayton, Ohio, specified in a contractual statement of work that ceramic coatings be further developed for metal foils. The H. I. Thompson Company of Los Angeles, California have from time to time requested samples of University of Illinois coatings as applied to such foil. They have supplied quantities of .002 inch thick sheets of Inconel, Nichrome, 18-8 stainless steels of the 347 and 321 types, and of carbon steel, described simply as "carbon steel." They are presently interested in protective coatings for Inconel, stainless-steel type 321, and carbon steel.

Selected coatings developed at the University of Illinois were applied to each of the metals. The coated specimens were tested for initial adherence, ability to protect the metal during extended heating at elevated temperatures and the adherence following such treatment. Coated specimens (4 inches by 4 inches) were furnished to the H. I. Thompson Company for further testing and evaluation. Samples of the coated foil, both before and after extended heating, are being prepared for submission to the Wright Air Development Center.

II. EXPERIMENTAL DETAILS

1. Metal Preparation

The nature of the metal foil made it impractical to use sandblasting as the method of metal preparation. Scouring the alloy metal surfaces with milled coating slip and rinsing in clear water was found to produce a surface which could be coated. It was later found that scouring the surface with coating slip or cleaning with either carbon tetrachloride or alcohol followed by heating for 10 minutes at 1800°F., in an electric furnace, produced a tightly adherent oxide film which aided in promoting and improving the adherence between the coating and the metal. This was particularly true for the Inconel and Nichrome foils which had polished surfaces. Pickling proved to be acceptable for the carbon steel foil and this method was used following standard practices with an alkali cleaner, hot sulphuric acid pickling batch, slightly acid nickel sulphate flashing solution and a borax-soda ash neutralizer.

2. Coating Preparation

Refractory ceramic coatings which were known to be durable at elevated temperatures and protective when applied to 0.050 inch thick sheets of Inconel, and the 18-8 grade stainless steels were selected for application to the foil rolled from these metals and also to that rolled from Nichrome. Coatings which were known to offer protection to iron, low carbon steel and Ti-Namel were prepared for testing on the carbon steel foil.

Coatings selected for application to the alloy metal foils were wet milled in 1000 gram quantities for a total of 10,000 revolutions in laboratory jar mills. This produced a desired reduction in particle size expressed as from 0.1 to 0.5 grams on a 325 mesh sieve from a 100 gram sample of coating slip. The coatings for carbon steel were milled to a fineness of from 0 to 1 gram on a 200 mesh sieve from a 100 gram sample of the slip.

The coatings prepared for application to the alloy metal foils included University of Illinois coatings No. 353-2, 353-5, 418-1, 418-3, 418-4, 418-7, and 423-2. Those made up for the protection of carbon steel foil were coatings No. 32-22, 285-1, 296-1, 327-1, 346-1, and 347-1. Of these, coatings No. 285-1, 296-1 and 327-1 were also applied to the stainless foil for purposes of comparison. The raw batch formulas and oxide compositions for these frits are presented in Table 1, the mill batch formulas in Table 2.

3. Coating Application

All coatings applied to the Inconel, Nichrome and stainless-steel foil were set to give a dry weight pick up of 13.5 to 14.5 grams per square foot, which is equivalent to a fired coating thickness of about 1.5 mils. The coatings for carbon steel were set to give a pick up of 18 to 20 grams per square foot equivalent to a fired thickness of about 2.0 mils.

The coatings were applied, by dipping, to 3-inch by 1-inch specimens cut from sheets of each of the metals. The individual specimens were numbered for identification.

All coatings were fired at the time and temperature which, by test, offered the best possibility of coverage and adherence. These firing times and temperatures are noted in Table 3 of this report.

4. Test Procedure

After firing, the coated specimens were observed for defects. Adherence was checked by flexing and by bending the corners of the specimens.

Coated and uncoated specimens of Inconel, Nichrome, and the stainless-steel foils (type 321 and 347) were hung on racks and subjected to an accelerated life test. This test was conducted at increasing temperatures. In the first complete cycle the specimens were heated for 24 hours at 1800°F. This included 16 hours at temperature together with an 8-hour period of cycling during which the samples were heated for 45 minutes at 1800°F. and cooled in air for 15 minutes. The alloy metal specimens were subjected to one complete cycle at 1800°F. followed by 16 hours of continuous heating at 1900°F. The specimens were then cooled to room temperature, observed for any evidence of deterioration of either the coating or the metal, and tested for coating adherence. The coated and uncoated specimens were compared for their relative resistance to oxidation.

Coated and uncoated specimens of the carbon-steel foil were tested following the same general procedure as outlined for the alloy metal foils. However, the test consisted of one complete 24-hour cycle at 1400°F., and one 24-hour cycle at 1500°F., followed by 16 hours of continuous heating at 1600°F.

Samples of the coated foil, as applied, and after test, are being prepared for shipment to the Power Plant Laboratory, Wright Air Development Center. The more protective coatings as determined for each of the metals were applied to other 4-inch by 4-inch specimens for submission to the H. I. Thompson Company.

III. RESULTS

1. Coating on Inconel

The application of coatings No. 285-1 and 418-1 to Inconel foil was not successful. Adherence did not develop and the coatings flaked off spontaneously upon cooling, leaving exposed bright metal. Coatings No. 353-5 and No. 418-3 produced a limited degree of adherence. The fired coating could be removed by flexing the metal severely. The initial adherence of coatings No. 296-1, 327-1, 353-2, 418-4, 418-7 and 423-2 was considered to be satisfactory in that it was necessary to deform the metal well past the elastic limit to effect a coating failure. The adherence of the No. 418 type coatings improved with increased chromic oxide content.

After the first 24 hours of the accelerated life test, at 1800°F., the adherence of all coatings tested showed a definite improvement. Coatings No. 296-1 and 327-1 had a tendency to burn back at the edges. Coatings No. 353-2, 353-5, 418-3, 418-4, 418-7 and 423-2 showed good protective properties with little change in appearance or quality except that the coatings became darker in color with a more pronounced matt surface texture. At this stage, coating No. 353-2 appeared to offer the best combination of desirable properties.

Continued heating of the specimens for 16 hours at 1900°F. resulted in further deterioration of the samples coated with coatings No. 296-1 and 327-1. The specimens coated with No. 423-1 became embrittled and broke when flexed. Coatings Nos. 353-2, 353-5, 418-3, 418-4, and 418-7 continued to offer protection, with coatings No. 418-4 and 418-7 being rated as the most protective.

A bare metal specimen subjected to the same heat-treatment oxidized to some extent with a resultant thinning down in cross-sectional area and a tendency to buckle and warp.

2. <u>Coatings on Nichrome</u>

The Nichrome foil, like the Inconel, had a polished surface which was not conducive to the promotion of adherence. Coatings No. 285-1, 418-1 and 418-3 did not adhere sufficiently to warrant further testing. Coatings No. 353-2 and 353-5 were weak in respect to adherence but could be handled and were subjected to extended heating. Coatings No. 296-1, 327-1, 418-4, 418-7 and 423-2 showed satisfactory adherence. As in the case with Inconel, increased chromic oxide content in the coating mill batch resulted in improved adherence.

Extended heating at 1800°F. together with cyclic heating and cooling improved the adherence between the metal and the coating. It resulted in a slight burn back at the edges of the samples coated with No. 296-1 and No. 327-1. Coatings No. 353-2, 353-5, 418-4, 418-7 and 423-2 evidenced good protective properties and good adherence. In every instance there was some change in color and in surface texture. All five coatings displayed matt surfaces even though No. 353-2 and 418-4 had been glossy prior to the heat treatment.

Heating at 1900°F. produced further noticeable deterioration of the specimens coated with No. 296-1 and 327-1. All of the other coatings continued to offer protection to the metal with little apparent change. Coatings No. 418-4 and 353-2 were rated the most protective with No. 353-5, 418-7 and 423-2 being considered as satisfactory coatings for the protection of Nichrome.

The bare metal showed some oxidation after heating at 1800°F. It was distorted, oxidized and reduced in cross-sectional area following the heating at 1900°F.

3. Coatings on 18-8 Stainless-Steel Type 321

All coatings developed better initial adherence when applied to stainless-steel type 321 than when applied to either Inconel or Nichrome, both of which had polished surfaces. Coating No. 285-1 was the only one tested in which the adherence was questionable. With this particular coating, a copperheading condition existed which left small areas protected by an oxide or slag rather than by the glass.

After the first 24 hours of testing at 1800°F., the copperheaded areas present in the No. 285-1 coating showed further oxidation. Coatings No. 296-1 and 327-1 were burned at the edges permitting oxidation of the metal at these points. Coatings of the 418 and 423 series were protective with satisfactory adherence. Of all the coatings tested, No. 353-2 and 353-5 were considered to show the best combination of protective properties and adherence.

Heating at 1900°F. was sufficiently severe to produce deterioration in all the coatings tested. This was evidenced by a burning back at the edges and a tendency for the test pieces to warp and buckle. Coatings No. 353-2, 353-5 and 418-1 were the least affected.

These results are very significant when it is pointed out that samples of the type 321 stainless steel, which were not protected by ceramic coatings, were completely oxidized after only 17 hours of heating at 1800°F.

4. Coatings on Stainless-Steel Type 347

In all instances the coatings applied to stainless-steel type 347 foil showed better adherence than they did on Inconel, Nichrome or 321 foil.

The first 24 hours of the accelerated life test at 1800°F. resulted in burning at the edges of the specimens coated with coatings No. 285-1, 296-1 and 327-1 although the protection and adherence in areas away from the edges was quite satisfactory. Coatings of the No. 353, 418 and 423 series showed good protective properties and an improvement in adherence.

Upon heating at 1900°F. coatings No. 285-1, 296-1 and 327-1 showed signs of deterioration as evidenced by the formation of copperheads, a burning back at the edges and distortion of the metal. Coatings No. 353-2, 353-5, 418-3, 418-4, 418-7 and 423-2, although protective, displayed a tendency to burn back at the edges.

Coating No. 418-1 however became quite glassy. It was protective up to and including the edges of the samples. It was considered to be the outstanding coating of those tested, for the protection of 18-8 stainless steel type 347.

Specimens of the bare metal subjected to the same progressive heat-treatment oxidized, warped, and retained less effective metal thickness than the coated specimens.

5. Coatings on Carbon Steel

All coatings tested produced continuous, adherent and protective coatings when applied to either scoured or pickled carbon steel foil.

Twenty-four hours of the accelerated life test at 1400°F. resulted in some burning at the edges for the samples coated with No. 346-1. Coatings 32-22, 285-1, 346-1 and 347-1 had a tendency to copperhead, but remained protective. At 1400°F. coatings No. 296-1 and 327-1 were the most effective.

An additional 24 hours of testing at 1500°F. produced little apparent change in the coated specimens except that coating No. 346-1 showed some tendency to burn back at the edges. Heating for an additional 16 hours at 1600°F. resulted in a burning back at the edges of all specimens. The adherence was reduced somewhat in all instances. Specimens coated with No. 296-1 and No. 327-1 were embrittled. Copperheading was noted for the specimens coated with No. 32-22, 285-1 and 347-1, but they did not become embrittled. Regardless of the copperheading condition, these coatings continued to protect the metal from severe oxidation that would destroy it completely after a relatively short exposure at temperatures above 1400°F.

Specimens of the bare metal oxidized very rapidly at temperatures as low as 1400°F., the metal being completely oxidized at the end of 17 hours of heating. The fact that appreciable metal remained in the coated specimens after 64 hours at temperatures ranging upward to 1600°F. proves the effectiveness of ceramic coatings in protecting metals against oxidation and prolonging their useful service life under severe operating conditions.

IV. DISCUSSION OF RESULTS

The results of this investigation show that ceramic coatings can prolong the useful service life of metal foil subjected to operation at elevated temperatures by protecting the metal against oxidation. The less resistant the alloy or metal is to oxidation the more apparent is the protection given by the ceramic coatings.

Methods of metal preparation offered some problem because of the thinness of the metal. Scouring the metal to remove all grease and dirt proved satisfactory in most cases. The production of a light, tightly adherent scale on the alloy metals, prior to coating, improved the adherence of the coating to the metal. Pickling was also shown to be satisfactory for the preparation of carbon-steel foil for coating.

The adherence of the fired coatings to the higher alloy content metals, Inconel and Nichrome, was somewhat lacking. This was considered to be due primarily to the polished surfaces since this was not noticeably true with the 18-8 type alloys which had an etched or satin-like finish. Increased chromic oxide in the mill resulted in a desirable increase in fired coating adherence. In every case, extended heating at 1800°F. resulted in improved adherence of coatings applied to alloy metal foil.

The fact that all coatings are not suitable for all metals is brought out by the results. A resume of the coatings[(1)] selected as the most protective for the various metal foils is presented as follows:

Metal	
1. Inconel	U. Ill. 418-4, U. Ill. 418-7
2. Nichrome	U. Ill. 418-4, U. Ill. 353-2
3. 321 type stainless steel	U. Ill. 353-2, U. Ill. 353-5, U. Ill 418-1
4. 347 type stainless steel	U. Ill. 418-1
5. Carbon steel	U. Ill. 32-22, U. Ill. 285-1, U. Ill. 347-1

[(1)] See Tables 1, 2, and 3 for frit compositions, mill batch formulas and firing schedules.

Coatings of the U. Ill. series 353, 418 and 423 were effective on the foil of the higher alloy content metals. However, coatings No. 418-4 and 418-7 were selected as the most protective for Inconel, while No. 353-2 and 418-4 were the best for application to Nichrome. Coatings No. 353-2, 353-5 and 418-1 were about equally effective when applied to 18-8 stainless steel type 321. Coating No. 418-1 proved to be outstanding when applied to stainless steel of the 347 type. Coatings of an altogether different composition were needed to produce satisfactory and protective coatings for carbon steel.

Table 1

Summary of Ceramic Coating Frit Compositions

Raw Batch Formula

Frit No.	32	285	296	327	346	347	353	418	423
Quartz	24.25	21.2	-	-	18.3	12.4	29.0	31.2	24.0
Potash Feldspar	34.80	30.2	85	83	47.4	59.9	-	-	-
Borax	23.80	21.0	-	-	17.9	11.9	-	-	-
Soda Ash	6.44	5.3	5	5	6.1	5.8	- -	-	-
Soda Nitre	4.13	4.0	5	5	4.4	4.6	-	-	-
Fluorspar	3.71	3.2	-	-	2.8	1.8	-	-	-
Cobalt Ox (Co_3O_4)	0.50	-	-	-	.37	0.25	-	-	-
Nickel Oxide (NiO)	0.50	-	-	-	.37	0.25	-	-	-
Manganese Dioxide	1.50	-	-	-	1.12	0.75	-	-	-
Aluminum Hydrate	-	15.1	-	-	-	-	-	-	-
Vanadium Pentoxide (90%)	-	-	5	5	1.25	2.5	-	-	3.5
Iron Oxide	-	-	-	2	-	-	-	-	-
Chromic Oxide	-	-	-	-	-	-	3.9	-	-
Boric Acid	-	-	-	-	-	-	11.1	12.0	24.1
Bismuth Nitrate	-	-	-	-	-	-	15.9	4.2	-
Calcium Carbonate	-	-	-	-	-	-	7.0	7.5	5.7
Cerium Oxide	-	-	-	-	-	-	3.9	4.2	4.0
Barium Carbonate	-	-	-	-	-	-	25.3	26.3	-
Zinc Oxide	-	-	-	-	-	-	3.9	4.2	4.0
Titanium Dioxide	-	-	-	-	-	-	-	4.2	12.0
Bismuth Trioxide	-	-	-	-	-	-	-	6.2	-
Strontium Carbonate	-	-	-	-	-	-	-	-	22.7
Total	99.63	100	100	100	100.01	100.15	100	100	100

Oxide Compositions

Frit No.	32	285	296	327	346	347	353	418	423
SiO_2	56.5	51.1	58.7	57.4	57.1	57.6	37.0	37.0	30.0
Al_2O_3	8.38	19.78	18.0	17.6	10.8	13.3	-	-	-
B_2O_3	10.55	9.55	-	-	8.0	5.3	8.0	8.0	17.0
Na_2O	12.06	10.88	7.3	7.3	10.9	9.8	-	-	-
K_2O	5.15	4.65	11.1	10.9	6.7	8.3	-	-	-
CaF_2	4.48	4.05	-	-	3.4	2.3	-	-	-
CoO	0.56	-	-	-	.45	0.3	-	-	-
NiO	0.60	-	-	-	.45	0.3	-	-	-
MnO_2	1.81	-	-	-	1.35	0.9	-	-	-
V_2O_5	-	-	4.9	4.9	1.2	1.85	-	-	4.0
Fe_2O_3	-	-	-	1.9	-	-	-	-	-
BiO_2	-	-	-	-	-	-	10.0	10.0	-
Cr_2O_3	-	-	-	-	-	-	5.0	-	-
CaO	-	-	-	-	-	-	5.0	5.0	4.0
BaO	-	-	-	-	-	-	25.0	25.0	-
ZnO	-	-	-	-	-	-	5.0	5.0	5.0
CeO_2	-	-	-	-	-	-	5.0	5.0	5.0
TiO_2	-	-	-	-	-	-	-	5.0	15.0
SrO									20.0
	100.09	100.01	100	100	100.35	99.95	100	100	100

Table 2

Summary of Mill Batch Compositions

1. Group A

Coating No.	353-2	353-5	418-1	418-3	418-4	418-7	423-2
Frit(1)	100	100	100	100	100	100	100
Cr_2O_3	20	-	-	-	25	40	-
TiO_2	-	10	-	10	-	-	15
Enamelers Clay	7	7	7	7	-	-	7
Clear Clay	-	-	-	-	6	6	-
Borax	0.75	-	0.75	-	-	-	-
Water	50	50	50	50	40	40	50
Fineness(2)	0-1	0-1	0-1	0-1	0-1	0-1	0-1

2. Group B

Coating No.	32-22	285-1	296-1	327-1	346-1	347-1
Frit(1)	88	100	100	100	100	100
Diaspore(3)	12	-	-	-	-	-
Enamelers Clay	7	7	7	7	7	7
Borax	0.75	0.75	0.75	0.75	0.75	0.75
Water	50	50	50	50	50	50
Fineness(4)	0-1	0-1	0-1	0-1	0-1	0-1

(1) The frit is designated by that portion of the coating number preceding the dash. The number following the dash refers to the mill batch formula. All frits preground to pass a 40-mesh sieve.

(2) Group A - Coatings were milled to a fineness of less than 1 gram retained on a 325-mesh sieve from a 100-gram sample.

(3) First Grade Diaspore, preground to pass 20-mesh sieve.

(4) Group B - Coatings were milled to a fineness of less than 1 gram retained on a 200-mesh sieve from a 100-gram sample of slip.

Table 3

Times and Firing Temperatures
for Various Coatings
As Applied to Metal Foil

Coating No.	Inconel		Nichrome		Stainless Steel Types 321		Stainless Steel Types 347		Carbon Steel	
	Time Min.	Temp. °F.	Time Min.	Temp. °F.	Time Min.	Temp °F.	Time Min.	Temp. °F.	Time Min.	Temp. °F.
353-2	6	1800	6	1800	6	1800	6	1800	-	-
353-5	6	1800	6	1800	6	1800	6	1800	-	-
418-1	5	1800	5	1800	5	1800	5	1800	-	-
418-3	6	1800	6	1800	6	1800	6	1800	-	-
418-4	6	1800	6	1800	6	1800	6	1800	-	-
418-7	6	1800	6	1800	6	1800	6	1800	-	-
423-2	6	1800	6	1850	6	1850	6	1800	-	-
32-22	-	-	-	-	-	-	-	-	4	1650
285-1	6	1800	6	1800	6	1800	6	1800	5	1750
296-1	6	2100	6	2100	6	2100	6	2100	$2\frac{1}{2}$	1950
327-1	6	2000	6	2000	6	2000	6	2000	$2\frac{1}{2}$	1900
346-1	-	-	-	-	-	-	-	-	4	1750
347-1	-	-	-	-	-	-	-	-	5	1750